

Appl. No. 10/667,134

Attorney Docket No. 89227.0005

Amdt. Dated June 19, 2006

Customer No. 26021

Reply to Advisory Action of March 16, 2006 and Office Action of October 20, 2005

Amendments to the Claims:

This listing of claims will replace all prior versions, and listings, of claims in the application:

-Listing of Claims:

1-2. (Canceled)

3. (Previously presented) The laminate as set forth in Claim 5, wherein:

the acid dianhydride component includes the pyromellitic dianhydride in a range of from 5 mole% to 90 mole%.

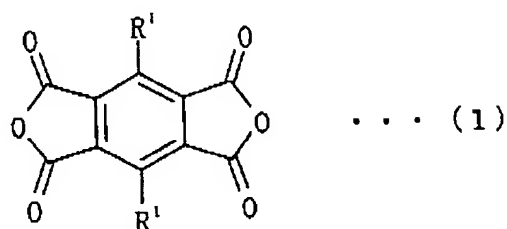
4. (Previously presented) The laminate as set forth in Claim 5, wherein:

the diamine component includes the paraphenylene diamine in a range of from 25 mole% to 75 mole%, and diaminodiphenyl ether in a range of from 25 mole% to 75 mole%.

5. (Currently amended) A laminate comprising a metal layer and a polyimide film, the metal layer being directly formed on the polyimide film having a dynamic viscoelasticity whose  $\tan \delta$  peak is located in a range of not less than 310°C but not more than 410°C, and whose  $\tan \delta$  value at 300°C is not more than 0.05, the polyimide film prepared by copolymerizing an acid dianhydride component and a diamine component,

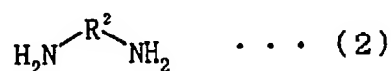
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the acid dianhydride component including a pyromellitic dianhydride represented by Equation (1):



where R¹ is a residue selected from a group consisting of H-, CH₃-, CF₃, Cl-, Br-, F-, and CH₃O-, and R¹ may be the same residues or different residues, and the diamine component including a paraphenylene diamine and a diaminodiphenyl ether,

the paraphenylene diamine being represented by Equation (2):



where R² is a bivalent aromatic group selected from a group consisting of:

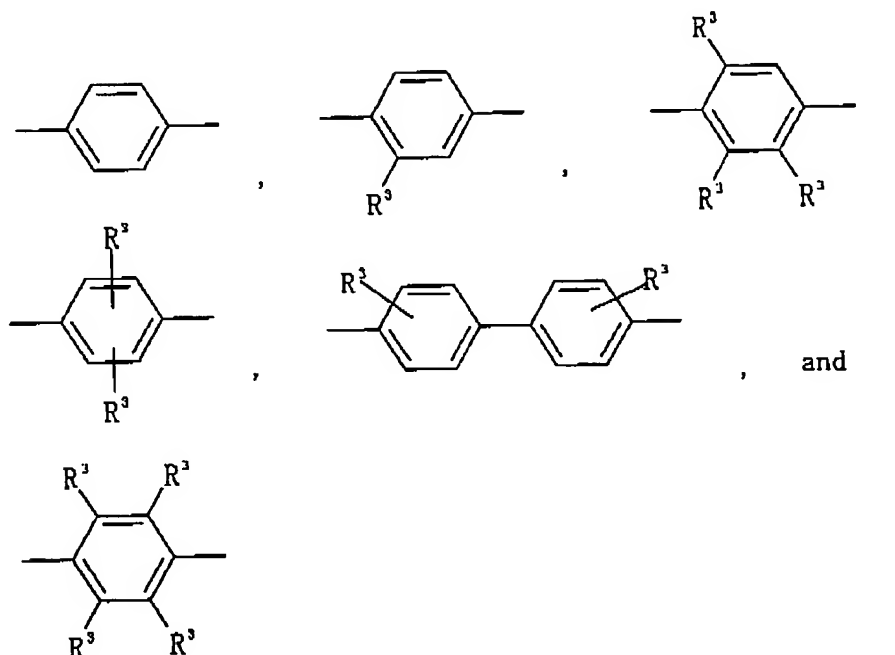
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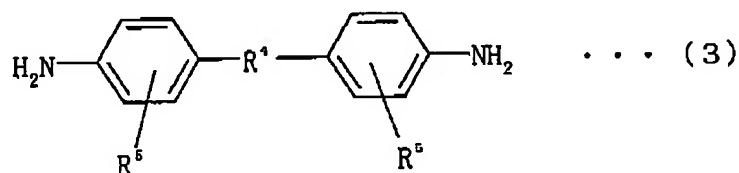
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and each R<sup>3</sup> in the group is independently any one of -H, -CH<sub>3</sub>, -OH, -CF<sub>3</sub>, -SO<sub>4</sub>, -COOH, -CO-NH<sub>2</sub>, -Cl, -Br, -F, and -OCH<sub>3</sub>, and the diaminodiphenyl ether being represented by General Formula (3):



where R<sup>4</sup> is a bivalent organic group selected from a group consisting of:

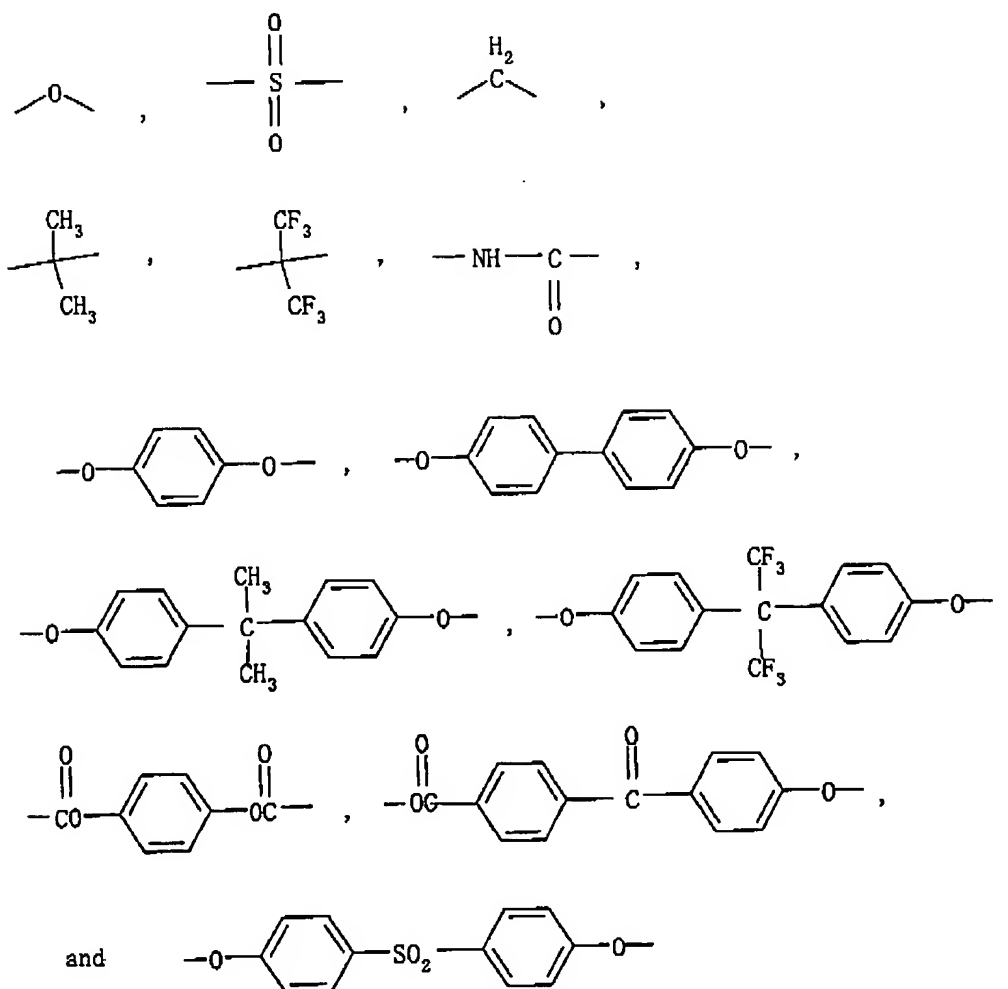
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and each R<sup>5</sup> in the group is independently any one of -H, -CH<sub>3</sub>, -OH, -CF<sub>3</sub>, -SO<sub>4</sub>, -COOH, -CO-NH<sub>2</sub>, -Cl, -Br, -F, and -OCH<sub>3</sub>,

wherein:

the acid dianhydride component further includes a bis(trimellitic monoester anhydride) and/or a biphenyl tetracarboxylic dianhydride,

the bis(trimellitic monoester anhydride) being represented by General Formula (4):

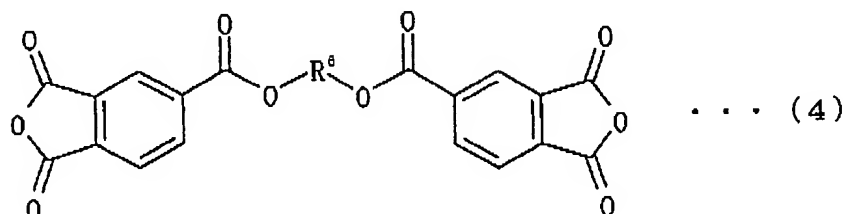
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where  $R^6$  is a bivalent organic group selected from a group consisting of:

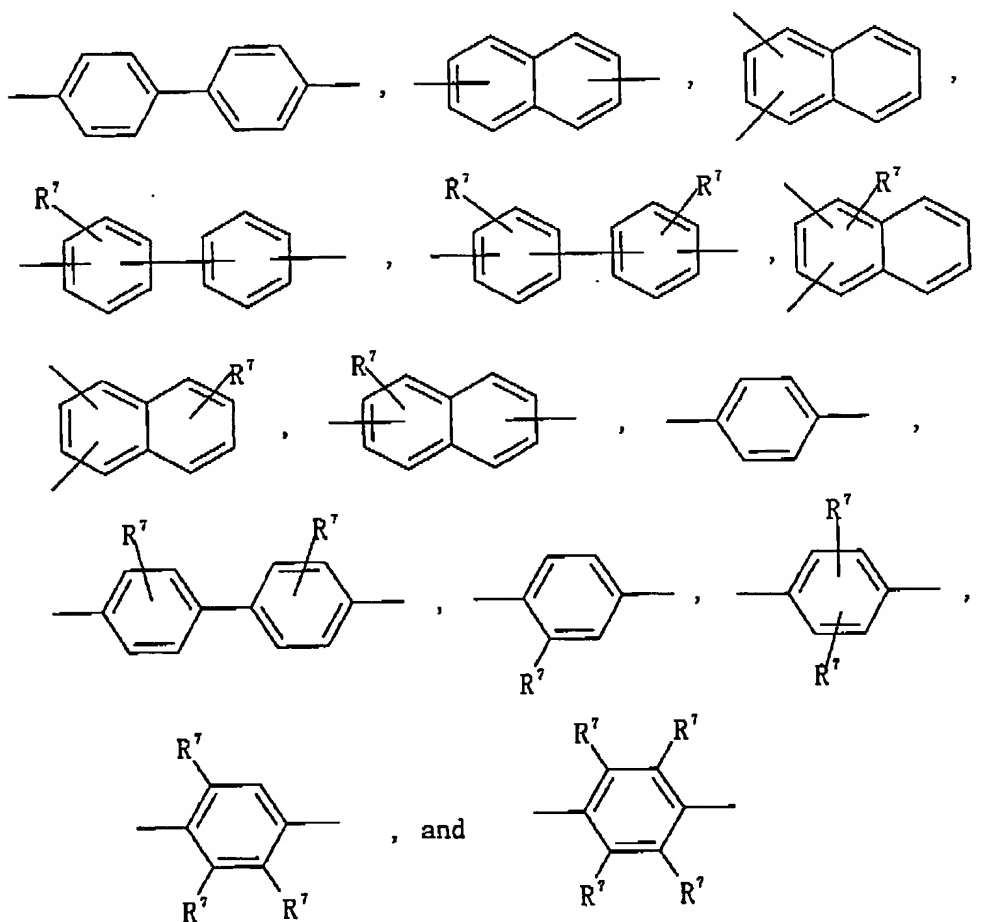
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and each R<sup>7</sup> is independently any one of -H, -CH<sub>3</sub>, -OH, -CF<sub>3</sub>, -SO<sub>4</sub>, -COOH, and -CO-NH<sub>2</sub>, and

~~the biphenyl tetracarboxylic dianhydride being represented by General Formula (5):~~

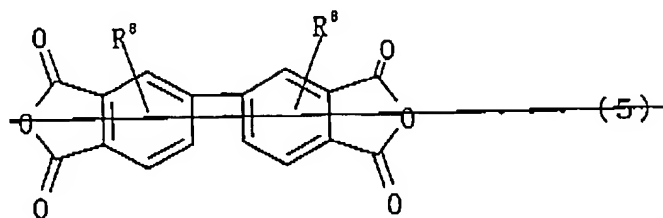
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~~where  $R^8$  is a residue selected from a group consisting of H,  $CH_3$ , Cl, Br, F and  $CH_3O$ , and  $R^8$  may be the same residues or the different residues.~~

6. (Previously presented) The laminate as set forth in Claim 5, wherein

the acid dianhydride component includes the bis(trimellitic monoester anhydride) in a range of from 20 mole% to 40 mole%.

7. (Previously presented) The laminate as set forth in Claim 5, wherein

the acid dianhydride component includes the biphenyl tetracarboxylic dianhydride in a range of from 0 mole% to 50 mole%.

8. (Previously presented) The laminate as set forth in Claim 5, wherein:

the polyimide film has a coefficient of hygroscopic expansion is 16ppm/%RH or less, and a water absorption percentage is 2.0% or less.

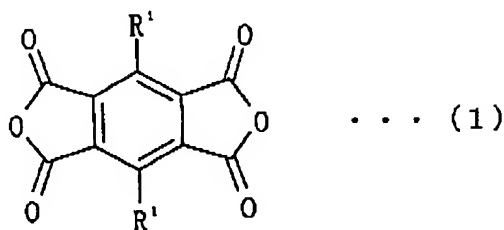
9-10. (Canceled)

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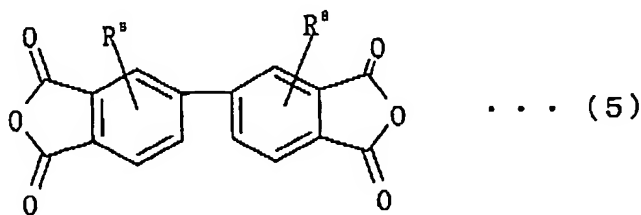
11. (Previously presented) A polyimide film prepared by copolymerizing an acid dianhydride component and a diamine component,

the acid dianhydride component including a pyromellitic dianhydride and a biphenyl tetracarboxylic dianhydride,

the pyromellitic dianhydride being represented by General Formula (1):



where R¹ is a residue selected from a group consisting of H-, CH₃-, CF₃-, Cl-, Br-, F-, and CH₃O-, and R¹ may be the same residues or different residues, and the biphenyl tetracarboxylic dianhydride being represented by General Formula (5):



where R² is a residue selected from a group consisting of H-, CH₃-, Cl-, Br-, F- and CH₃O-, and R² may be the same residues or the different residues, and



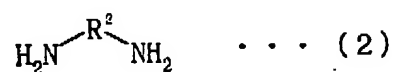
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the polyimide film having such an etching speed that one side thereof is etched with a 1N potassium hydroxide solution at an etching speed of 0.1 $\mu$ m/minute (one side) or higher.

12. (Previously presented) The polyimide film as set forth in Claim 11, wherein:

the diamine component includes a paraphenylene diamine and/or a diaminodiphenyl ether,

the paraphenylene diamine being represented by General Formula (2):



where R<sup>2</sup> is a bivalent aromatic group selected from a group consisting of:

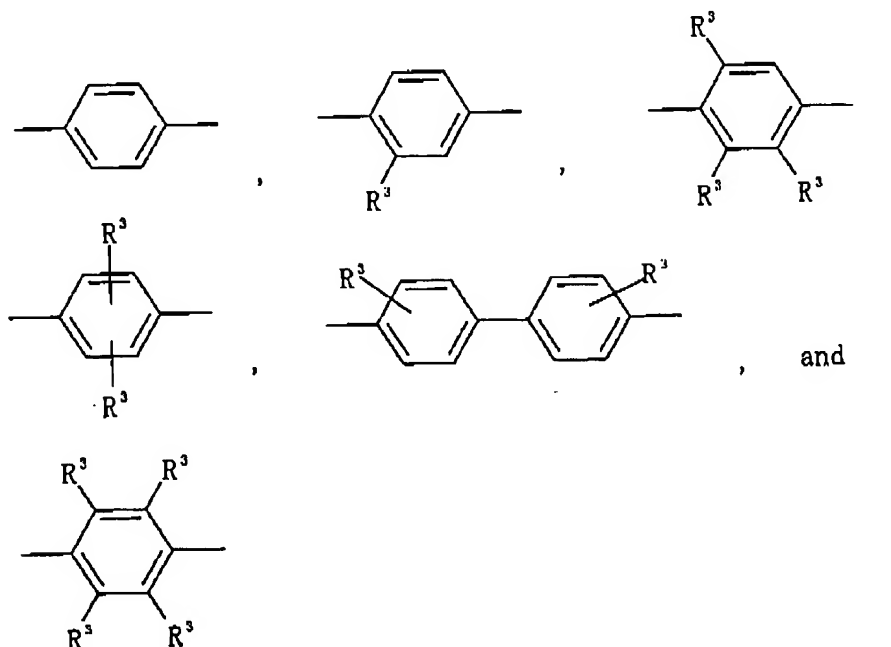
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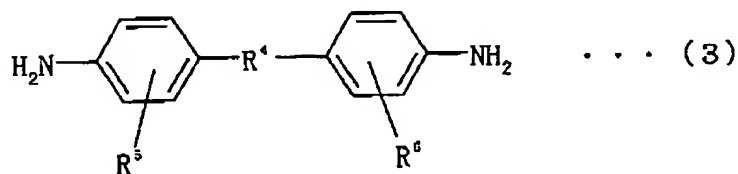
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and each  $R^3$  in the group is independently any one of  $-H$ ,  $-CH_3$ ,  $-OH$ ,  $-CF_3$ ,  $-SO_4$ ,  $-COOH$ ,  $-CO-NH_2$ ,  $-Cl$ ,  $-Br$ ,  $-F$ , and  $-OCH_3$ , and the diaminodiphenyl ether being represented by General Formula (3):



where  $R^4$  is a bivalent organic group selected from a group consisting of:

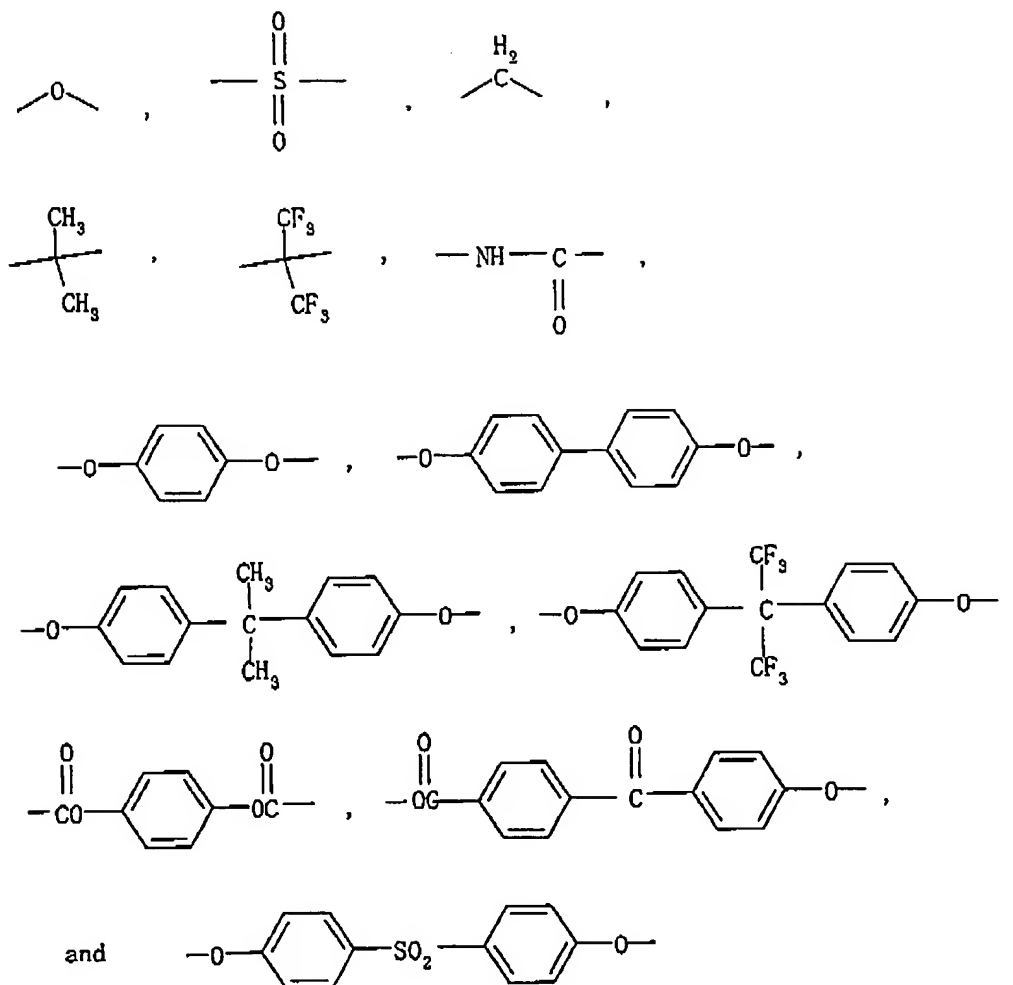
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and each R<sup>5</sup> in the group is independently any one of -H, -CH<sub>3</sub>, -OH, -CF<sub>3</sub>, -SO<sub>4</sub>, -COOH, -CO-NH<sub>2</sub>, -Cl, -Br, -F, and -OCH<sub>3</sub>.

13. (Original) The polyimide film as set forth in Claim 11, wherein:

the acid dianhydride component includes the pyromellitic dianhydride in a range of from 30 mole% to 99.9 mole%, and the

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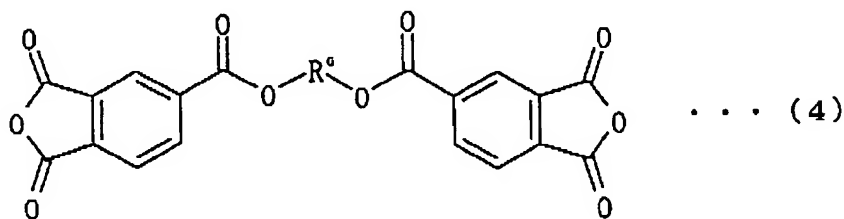
biphenyl tetracarboxylic dianhydride in a range of from 0.1 mole% to 50 mole%.

14. (Original) The polyimide film as set forth in Claim 12, wherein:

the diamine component includes the paraphenylene diamine in a range of from 15 mole% to 85 mole%, and diaminodiphenyl ether in a range of from 15 mole% to 85 mole%.

15. (Previously presented) The polyimide film as set forth in Claim 12,

the acid dianhydride component further includes a bis(trimellitic monoester anhydride) being represented by General Formula (4):



where  $R^6$  is a bivalent organic group selected from a group consisting of:

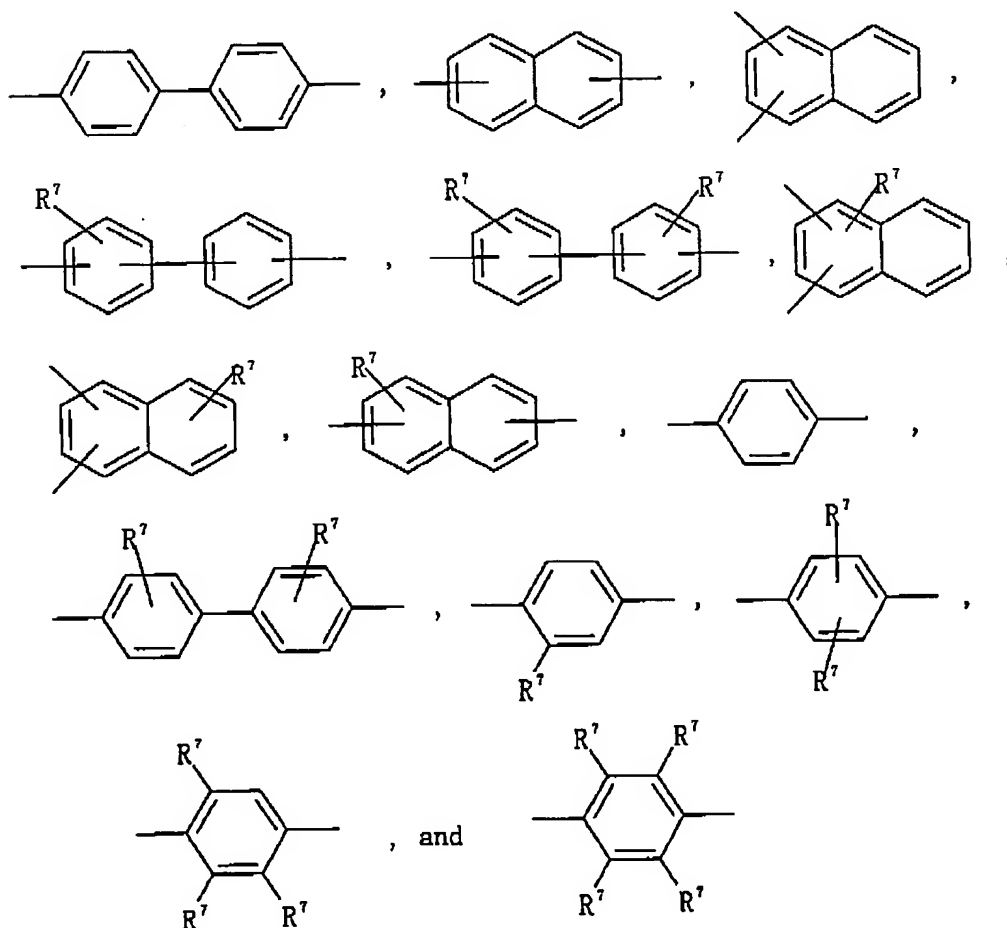
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and each R<sup>7</sup> is independently any one of -H, -CH<sub>3</sub>, -OH, -CF<sub>3</sub>, -SO<sub>4</sub>, -COOH, and -CO-NH<sub>2</sub>.

16. (Original) The polyimide film as set forth in Claim 15, wherein:

The acid dianhydride component includes the bis(trimellitic monoester anhydride) in a range of from 10 mole% to 50 mole%.

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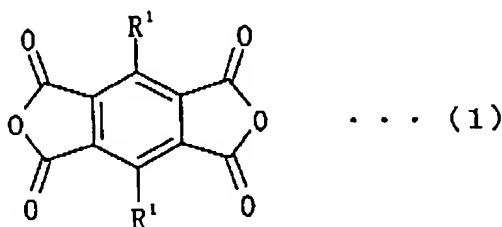
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17. (Original) The polyimide film as set forth in Claim 11, wherein:

a retention percent of tear-through resistance of the polyimide film after exposing the polyimide film to environment of a temperature of 150°C, a humidity of 100%RH, and 4 atmospheric pressure for 48 hours is not less than 50%.

18. (Currently amended) ~~Laminate, comprising:~~  
~~a metal layer; and~~ A laminate that is manufactured with  
a polyimide film that is prepared by copolymerizing an acid dianhydride component and a diamine component,  
the acid dianhydride component including a pyromellitic dianhydride and a biphenyl tetracarboxylic dianhydride, the pyromellitic dianhydride being represented by General Formula (1):



where R¹ is a residue selected from a group consisting of H-, CH₃-, CF₃, Cl-, Br-, F-, and CH₃O-, and R¹ may be the same residues or different residues, and

the a biphenyl tetracarboxylic dianhydride being represented by General Formula (5):

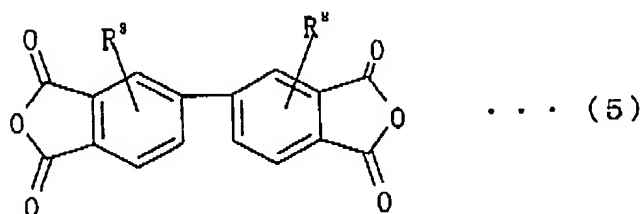
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where  $R^s$  is a residue selected from a group consisting of H-,  $\text{CH}_3$ -, Cl-, Br-, F- and  $\text{CH}_3\text{O}$ -, and  $R^s$  may be the same residues or the different residues, and

the diamine component including a paraphenylene diamine represented by General Formula (2)



where  $R^2$  is a bivalent aromatic group selected from a group consisting of:





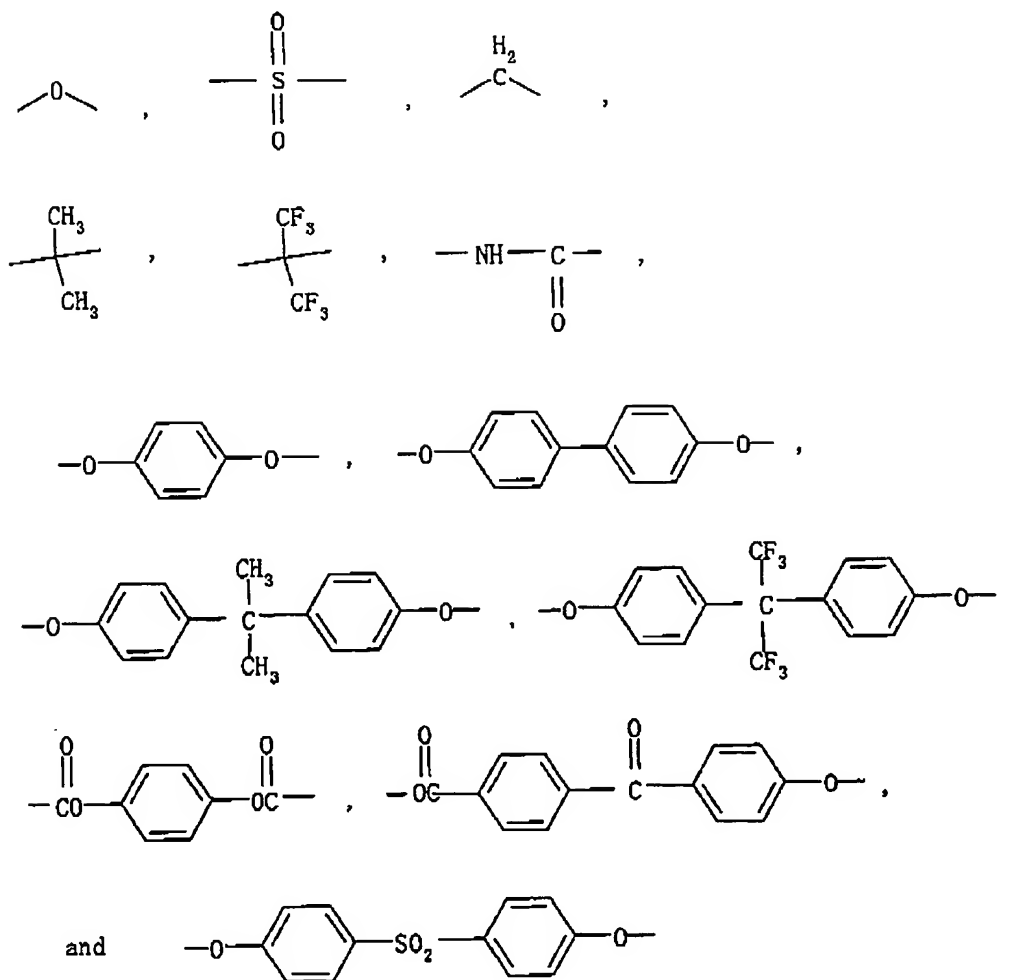
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and each R<sup>5</sup> in the group is independently any one of -H, -CH<sub>3</sub>, -OH, -CF<sub>3</sub>, -SO<sub>4</sub>, -COOH, -CO-NH<sub>2</sub>, -Cl, -Br, -F, and -OCH<sub>3</sub>,

wherein:

the acid dianhydride component further including a bis(trimellitic monoester anhydride) represented by General Formula (4).

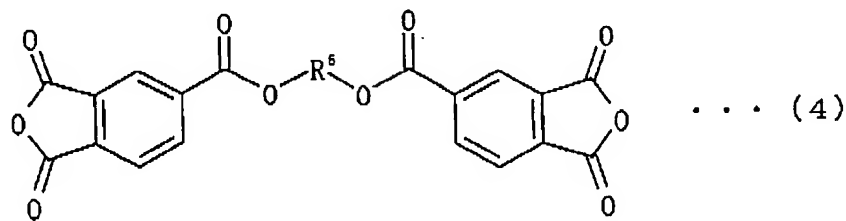
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where R<sup>6</sup> is a bivalent organic group selected from a group consisting of:



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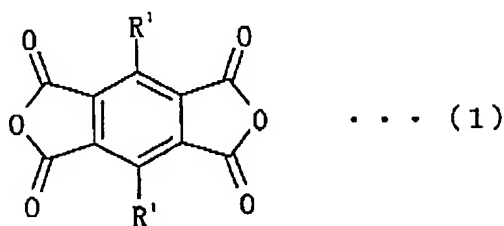
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19. (Previously presented) A polyimide film prepared by copolymerizing an acid dianhydride component and a diamine component,

the acid dianhydride component including the pyromellitic dianhydride, represented by General Formula (1), in a range of from 40 mole% to 80 mole%, the biphenyl tetracarboxylic dianhydride, represented by General Formula (5) in a range of from 1 mole% to 40 mole%, and the bis(trimellitic monoester anhydride, represented by General Formula (4), in a range of from 20 mole% to 50 mole%, and

the diamine component including the paraphenylene diamine, represented by General Formula (2), in a range of 25 mole% to 75 mole%, and the diaminodiphenyl ether, represented by General Formula (3), in a range of 25 mole% to 75 mole%, where General Formula (1) is:



where R¹ is a residue selected from a group consisting of H-, CH₃-, CF₃, Cl-, Br-, F-, and CH₃O-, and R¹ may be the same residues or different residues;

General Formula (5) is:

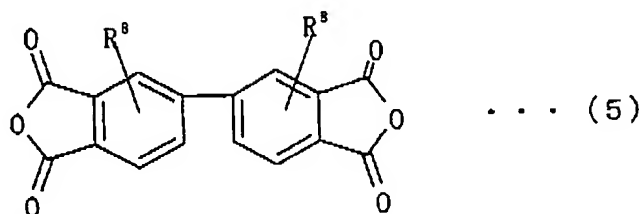
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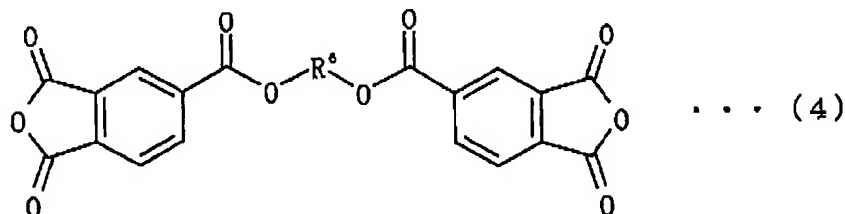
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where  $R^8$  is a residue selected from a group consisting of H-,  $CH_3$ -, Cl-, Br-, F- and  $CH_3O$ -, and  $R^8$  may be the same residues or the different residues;

General Formula (4) is:



where  $R^6$  is a bivalent organic group selected from a group consisting of:

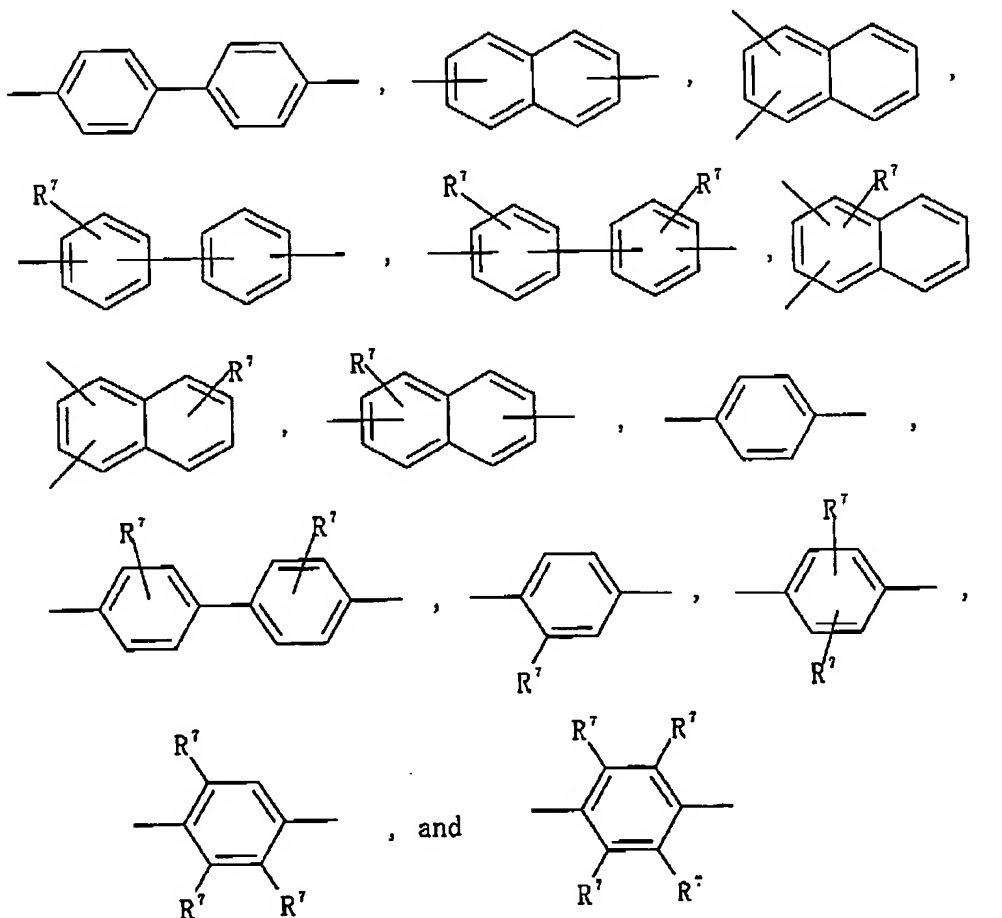
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and each R<sup>7</sup> is independently any one of -H, -CH<sub>3</sub>, -OH, -CF<sub>3</sub>, -SO<sub>4</sub>, -COOH, and -CO-NH<sub>2</sub>;

General Formula (2) is:



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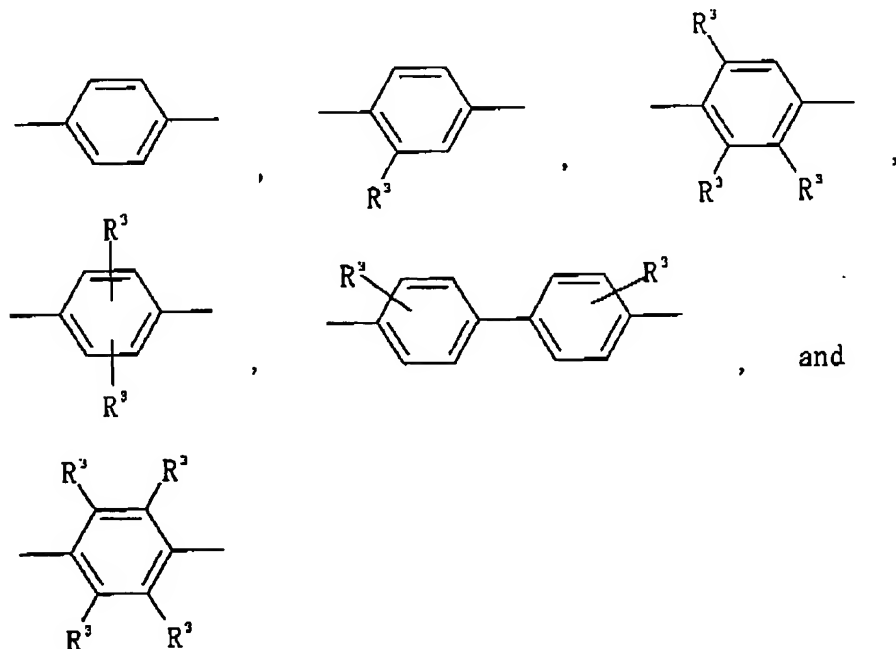
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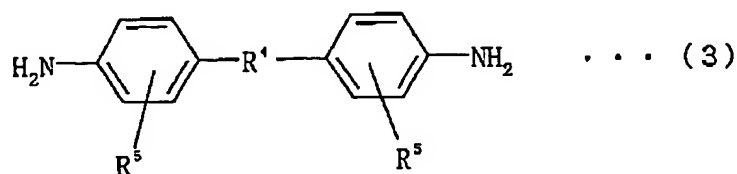
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where  $R^2$  is a bivalent aromatic group selected from a group consisting of:



and each  $R^3$  in the group is independently any one of -H, -CH<sub>3</sub>, -OH, -CF<sub>3</sub>, -SO<sub>4</sub>, -COOH, -CO-NH<sub>2</sub>, -Cl, -Br, -F, and -OCH<sub>3</sub>; and

General Formula (3) is:



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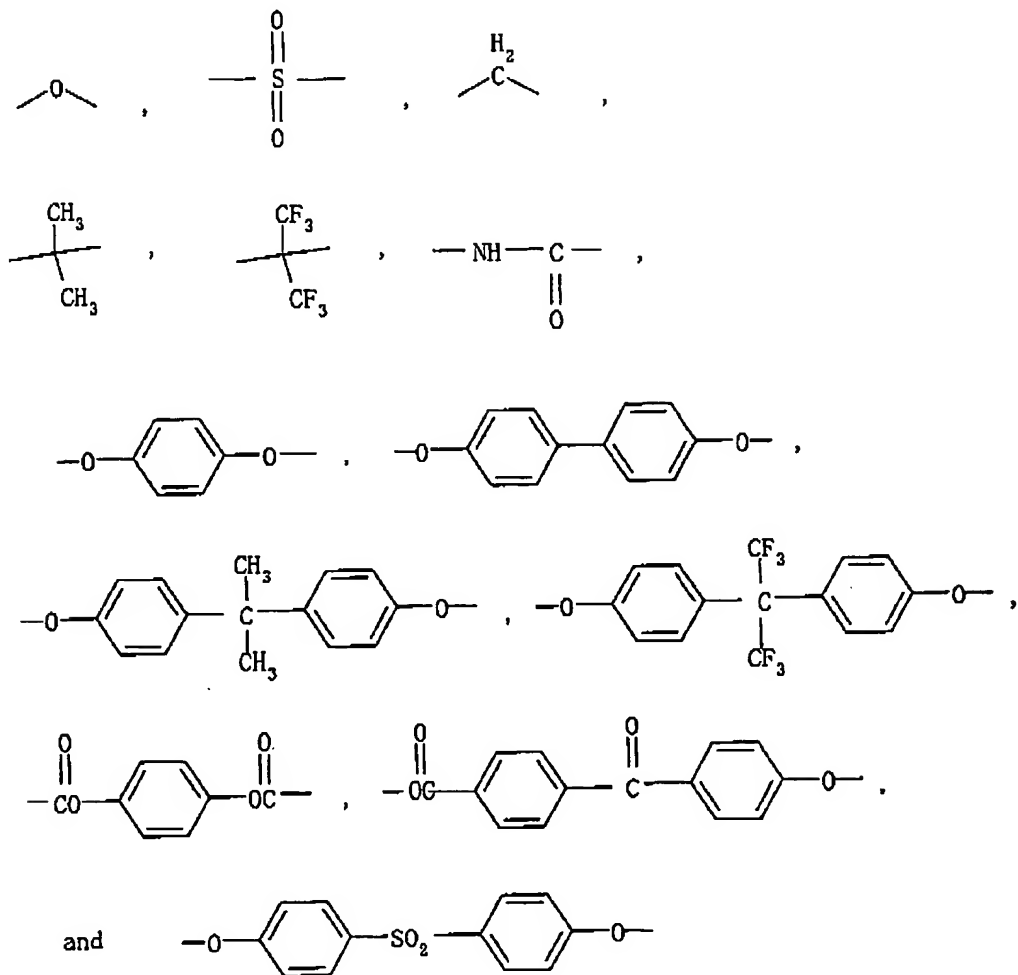
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where R<sup>4</sup> is a bivalent organic group selected from a group consisting of:



and each R<sup>5</sup> in the group is independently any one of -H, -CH<sub>3</sub>, -OH, -CF<sub>3</sub>, -SO<sub>4</sub>, -COOH, -CO-NH<sub>2</sub>, -Cl, -Br, -F, and -OCH<sub>3</sub>.



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20. (Original) The polyimide film as set forth in Claim 19, the polyimide film having a thickness in a range of from  $1\mu\text{m}$  to  $200\mu\text{m}$ .

21. (Original) The polyimide film as set forth in Claim 19, the polyimide film having a modulus of elasticity in a range of from  $500\text{kg/mm}^2$  to  $800\text{kg/mm}^2$ .

22. (Original) The polyimide film as set forth in Claim 19, the polyimide film having a coefficient of hygroscopic expansion in a range of from  $2\text{ppm}/\%RH$  to  $20\text{ppm}/\%RH$ .

23. (Original) The polyimide film as set forth in Claim 19, the polyimide film having a coefficient of liner expansion in a range of 1 to  $30 \times 10^{-6}\text{cm/cm}/^{\circ}\text{C}$  at a temperature of from  $100^{\circ}\text{C}$  to  $200^{\circ}\text{C}$ .

24. (Original) The polyimide film as set forth in Claim 19, wherein:

a peel strength at an interface between the polyimide film and a metal layer of laminate is not less than  $5\text{N/cm}$ , the laminate having the polyimide film and the metal layer that is formed on the polyimide film by vacuum depositing and electroplating; and

a retention rate of the peel strength is not less than 10% after exposing the laminate to environment of a temperature of  $121^{\circ}\text{C}$  and a humidity of 100%RH for 12 hours.

25. (Previously presented) Laminate comprising:  
a metal layer; and

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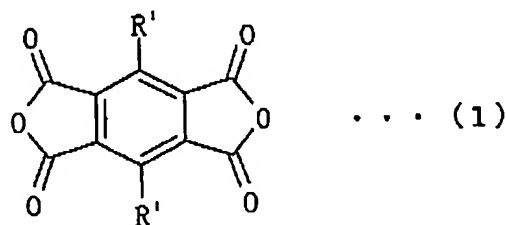
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a polyimide film prepared by copolymerizing an acid dianhydride component and a diamine component,

the acid dianhydride component including the pyromellitic dianhydride, represented by General Formula (1), in a range of from 40 mole% to 80 mole%, the biphenyl tetracarboxylic dianhydride, represented by General Formula (5) in a range of from 1 mole% to 40 mole%, and the bis(trimellitic monoester anhydride), represented by General Formula (4), in a range of from 20 mole% to 50 mole%, and

the diamine component including the paraphenylene diamine, represented by General Formula (2), in a range of 25 mole% to 75 mole%, and the diaminodiphenyl ether, represented by General Formula (3), in a range of 25 mole% to 75 mole%, where General Formula (1) is:



where  $R^1$  is a residue selected from a group consisting of H-,  $CH_3$ -,  $CF_3$ , Cl-, Br-, F-, and  $CH_3O$ -, and  $R^1$  may be the same residues or different residues;

General Formula (5) is:

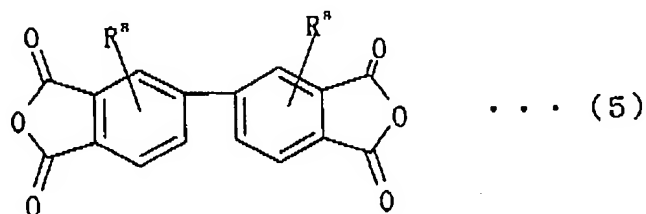
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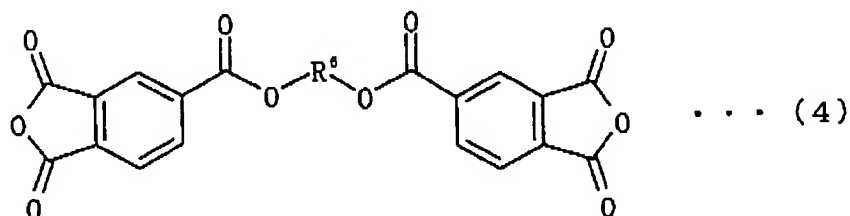
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where  $R^8$  is a residue selected from a group consisting of H-,  $CH_3$ -, Cl-, Br-, F- and  $CH_3O$ -, and  $R^8$  may be the same residues or the different residues;

General Formula (4) is:



where  $R^6$  is a bivalent organic group selected from a group consisting of:

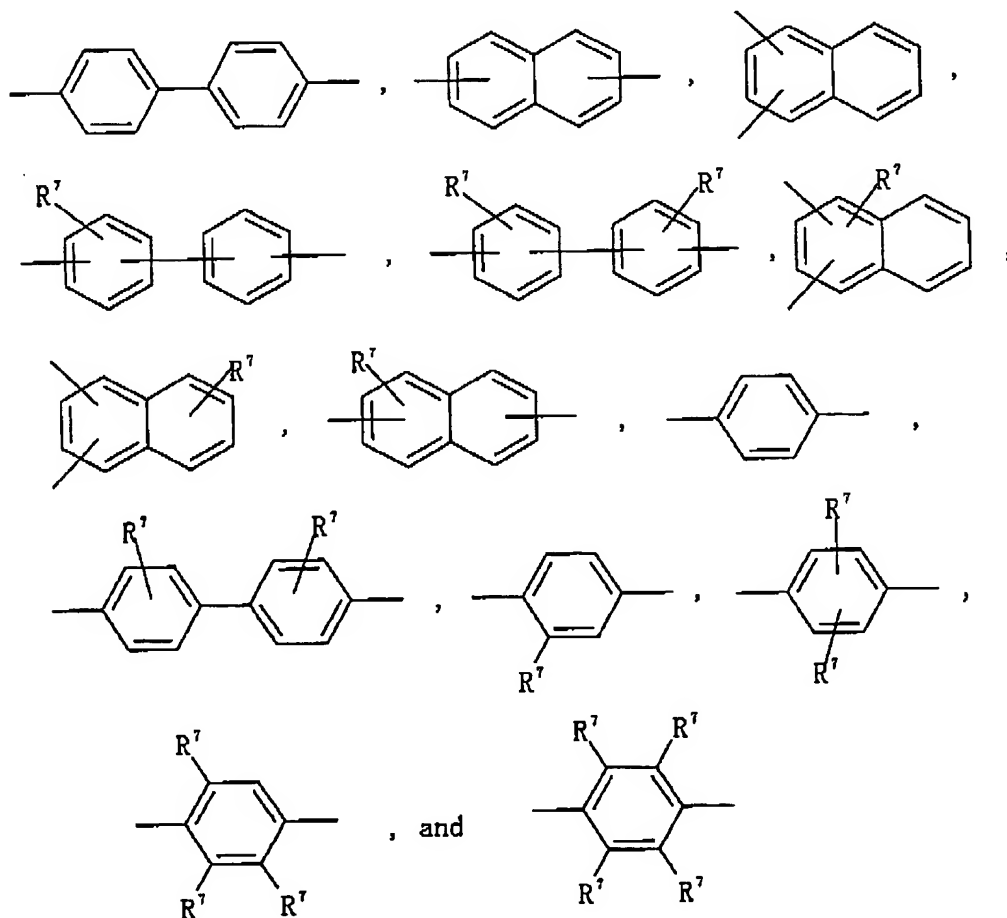
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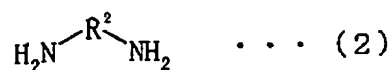
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and each  $R^7$  is independently any one of  $-H$ ,  $-CH_3$ ,  $-OH$ ,  $-CF_3$ ,  $-SO_4$ ,  $-COOH$ , and  $-CO-NH_2$ ;

General Formula (2) is:

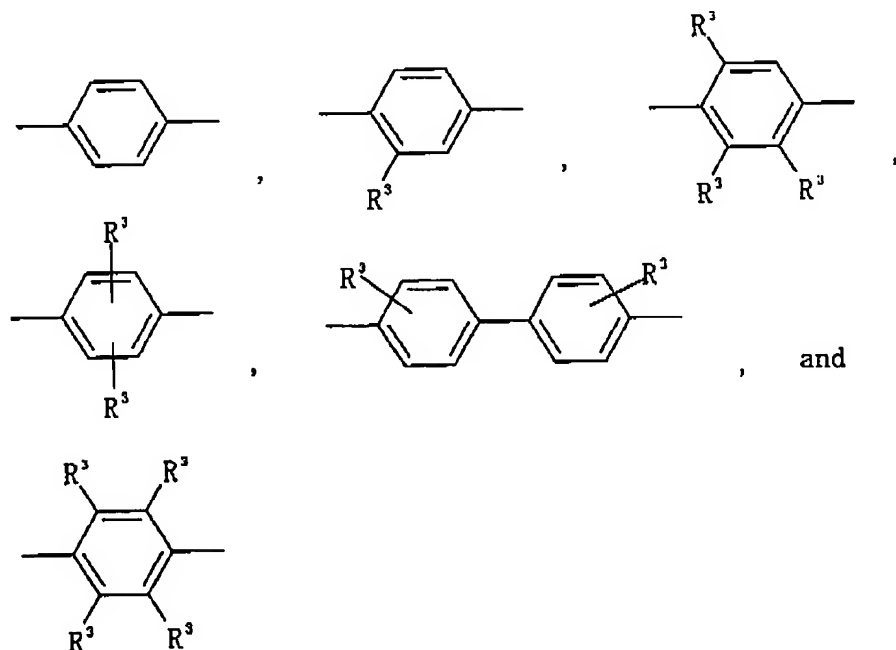


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Attorney Docket No. 89227.0005

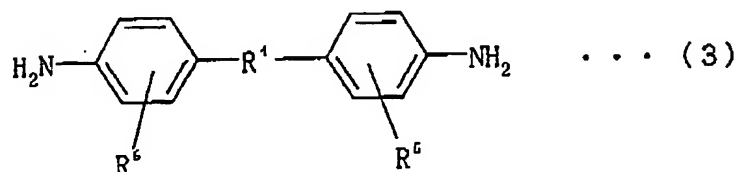
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where  $R^2$  is a bivalent aromatic group selected from a group consisting of:



and each  $R^3$  in the group is independently any one of -H, -CH<sub>3</sub>, -OH, -CF<sub>3</sub>, -SO<sub>4</sub>, -COOH, -CO-NH<sub>2</sub>, -Cl, -Br, -F, and -OCH<sub>3</sub>; and

General Formula (3) is:



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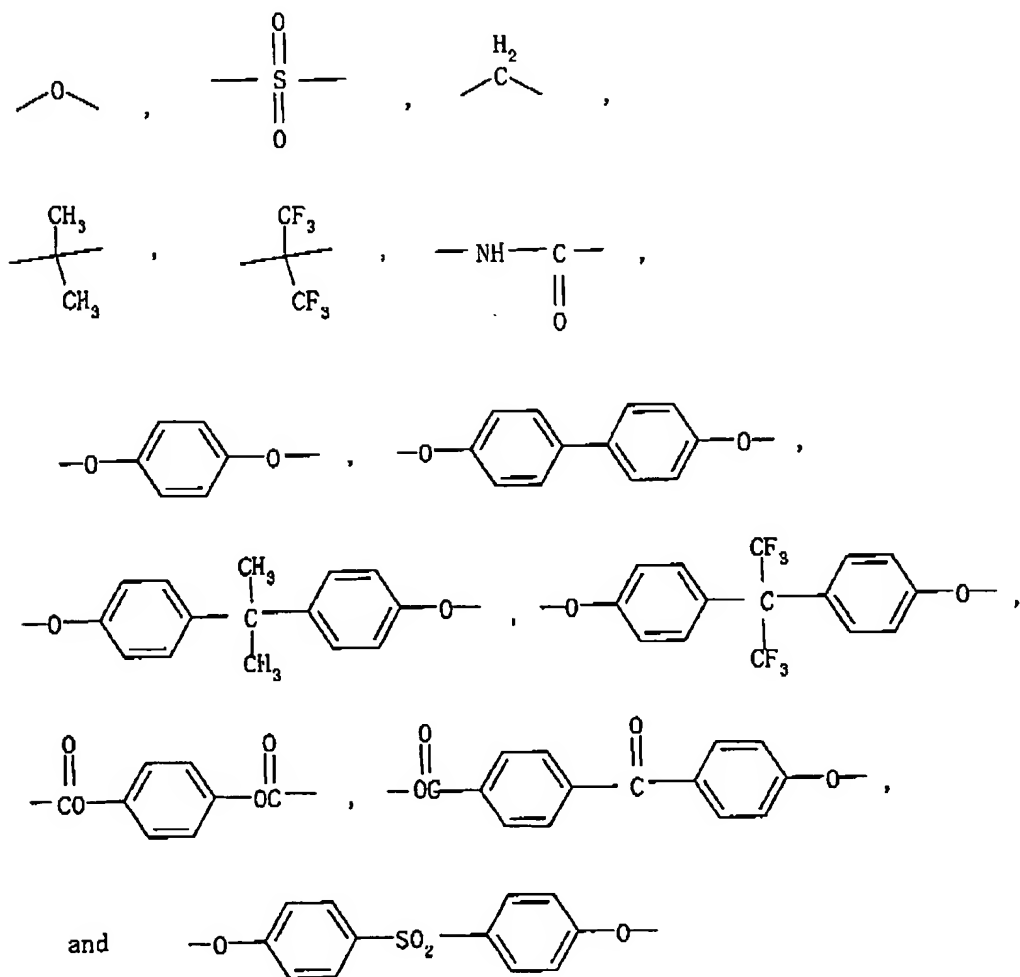
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where R<sup>4</sup> is a bivalent organic group selected from a group consisting of:



and each R<sup>5</sup> in the group is independently any one of -H, -CH<sub>3</sub>, -OH, -CF<sub>3</sub>, -SO<sub>4</sub>, -COOH, -CO-NH<sub>2</sub>, -Cl, -Br, -F, and -OCH<sub>3</sub>.

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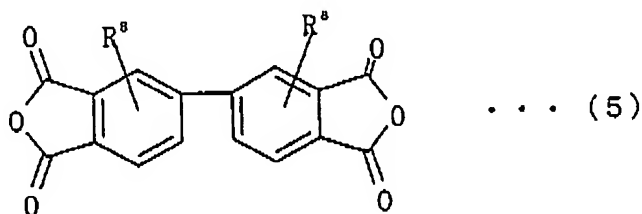
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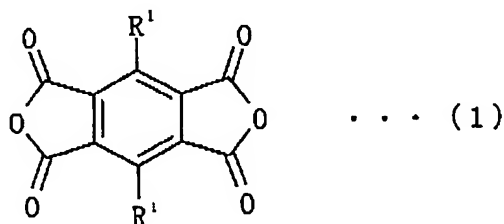
26. (New) The laminate as set forth in claim 5, wherein the acid dianhydride component includes a biphenyl tetracarboxylic dianhydride represented by General Formula (5)



where  $R^8$  is a residue selected from a group consisting of H-,  $CH_3$ -, Cl-, Br-, F- and  $CH_3O$ -, and  $R^8$  may be the same residues or the different residues.

27. (New) A polyimide film prepared by copolymerizing an acid dianhydride component and a diamine component,

the acid dianhydride component including a pyromellitic dianhydride represented by General Formula (1),



where  $R^1$  is a residue selected from a group consisting of H-,  $CH_3$ -,  $CF_3$ -, Cl-, Br-, F-, and  $CH_3O$ -, and  $R^1$  may be the same residues or different residues, and

the diamine component including a paraphenylene diamine represented by General Formula (2)

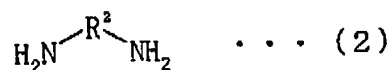
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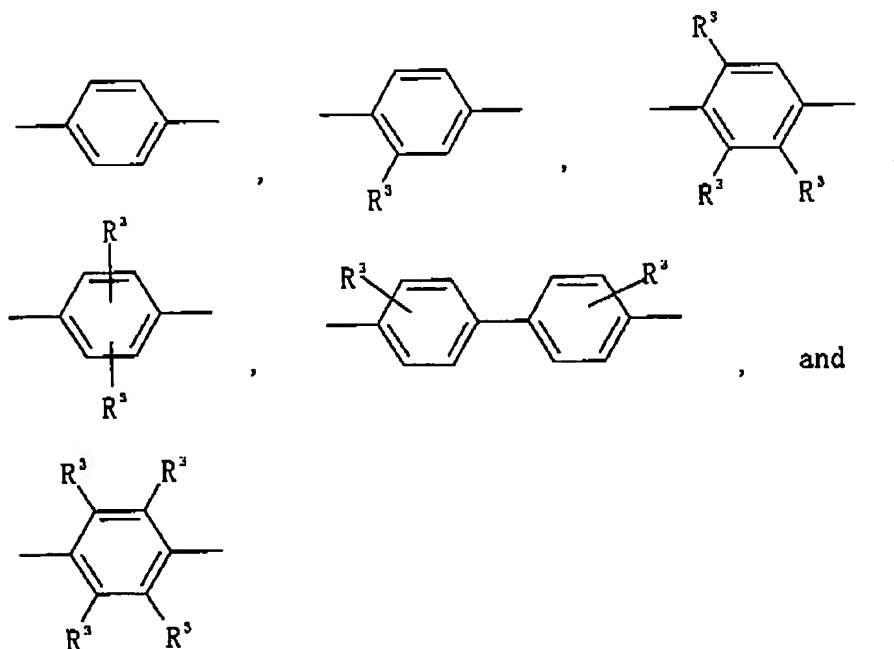
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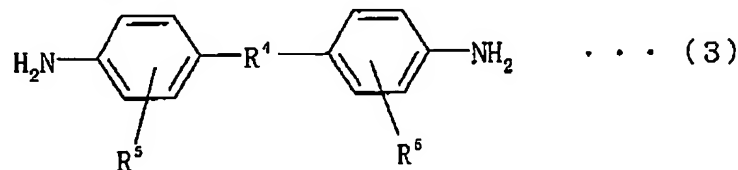
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where  $\text{R}^2$  is a bivalent aromatic group selected from a group consisting of:



and each  $\text{R}^3$  in the group is independently any one of -H, -CH<sub>3</sub>, -OH, -CF<sub>3</sub>, -SO<sub>4</sub>, -COOH, -CO-NH<sub>2</sub>, -Cl, -Br, -F, and -OCH<sub>3</sub>, and a diaminodiphenyl ether represented by General Formula (3),





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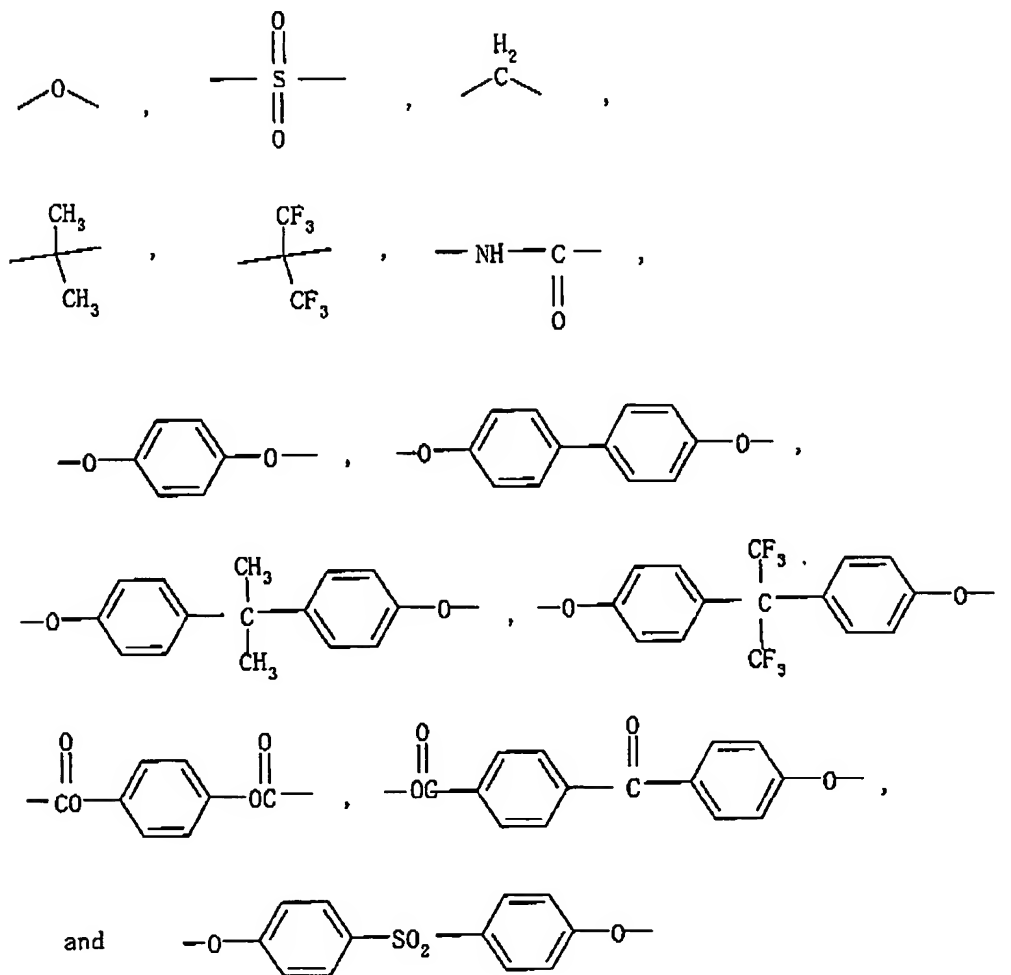
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where  $R^4$  is a bivalent organic group selected from a group consisting of:



and each  $R^5$  in the group is independently any one of -H, -CH<sub>3</sub>, -OH, -CF<sub>3</sub>, -SO<sub>4</sub>, -COOH, -CO-NH<sub>2</sub>, -Cl, -Br, -F, and -OCH<sub>3</sub>,

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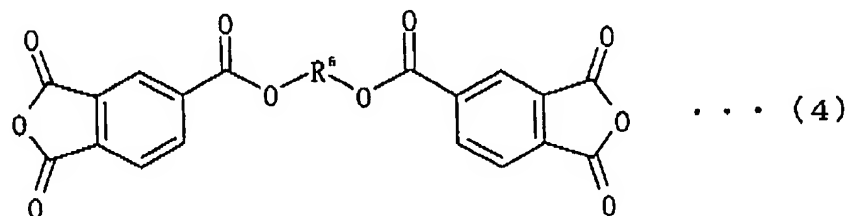
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wherein:

the acid dianhydride component further including a bis(trimellitic monoester anhydride) represented by General Formula (4),



where  $R^6$  is a bivalent organic group selected from a group consisting of:

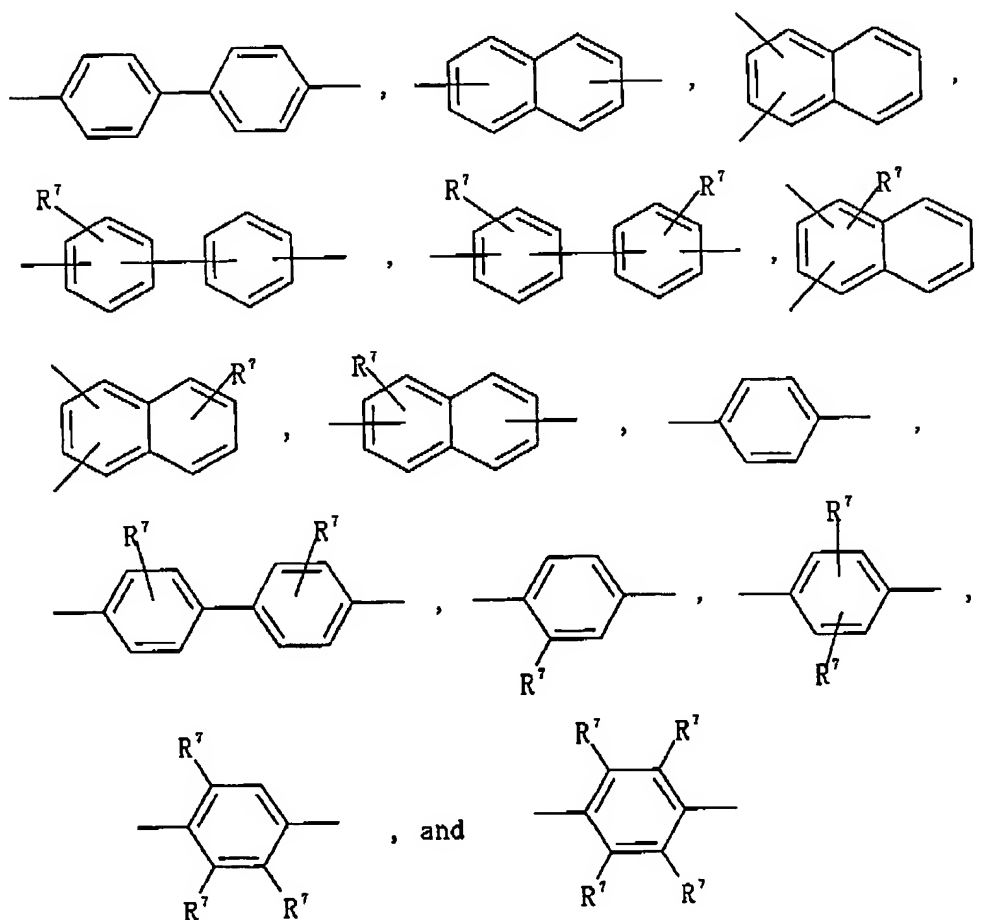
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and each R<sup>7</sup> is independently any one of -H, -CH<sub>3</sub>, -OH, -CF<sub>3</sub>, -SO<sub>4</sub>, -COOH, and -CO-NH<sub>2</sub>,

the polyimide film having a dynamic viscoelasticity whose tan δ peak is located in a range of not less than 310°C but not more than 410°C, and whose tan δ value at 300°C is not more than 0.05.

28. (New) A polyimide film prepared by copolymerizing an acid dianhydride component and a diamine component,

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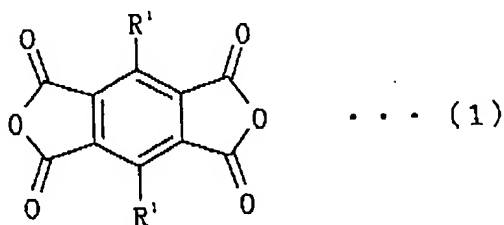
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the acid dianhydride component including a pyromellitic dianhydride represented by General Formula (1),



where R¹ is a residue selected from a group consisting of H-, CH₃-, CF₃-, Cl-, Br-, F-, and CH₃O-, and R¹ may be the same residues or different residues, and

the diamine component including a paraphenylene diamine represented by General Formula (2)



where R² is a bivalent aromatic group selected from a group consisting of:



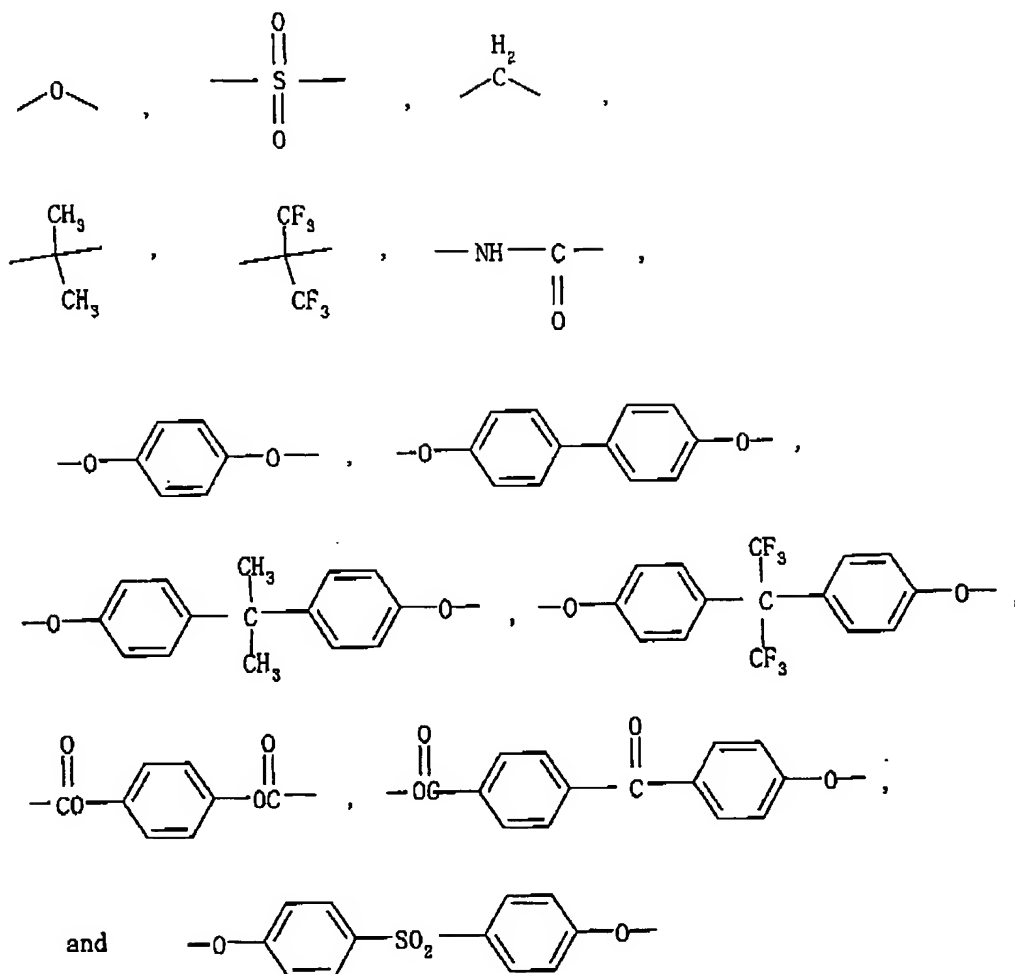
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and each R<sup>5</sup> in the group is independently any one of -H, -CH<sub>3</sub>, -OH, -CF<sub>3</sub>, -SO<sub>4</sub>, -COOH, -CO-NH<sub>2</sub>, -Cl, -Br, -F, and -OCH<sub>3</sub>,  
wherein:

the acid dianhydride component further including a bis(trimellitic monoester anhydride) represented by General Formula (4),

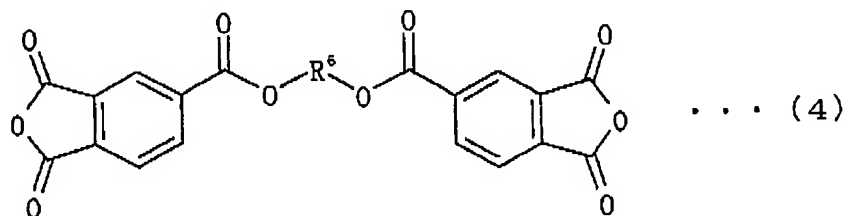
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where  $R^6$  is a bivalent organic group selected from a group consisting of:

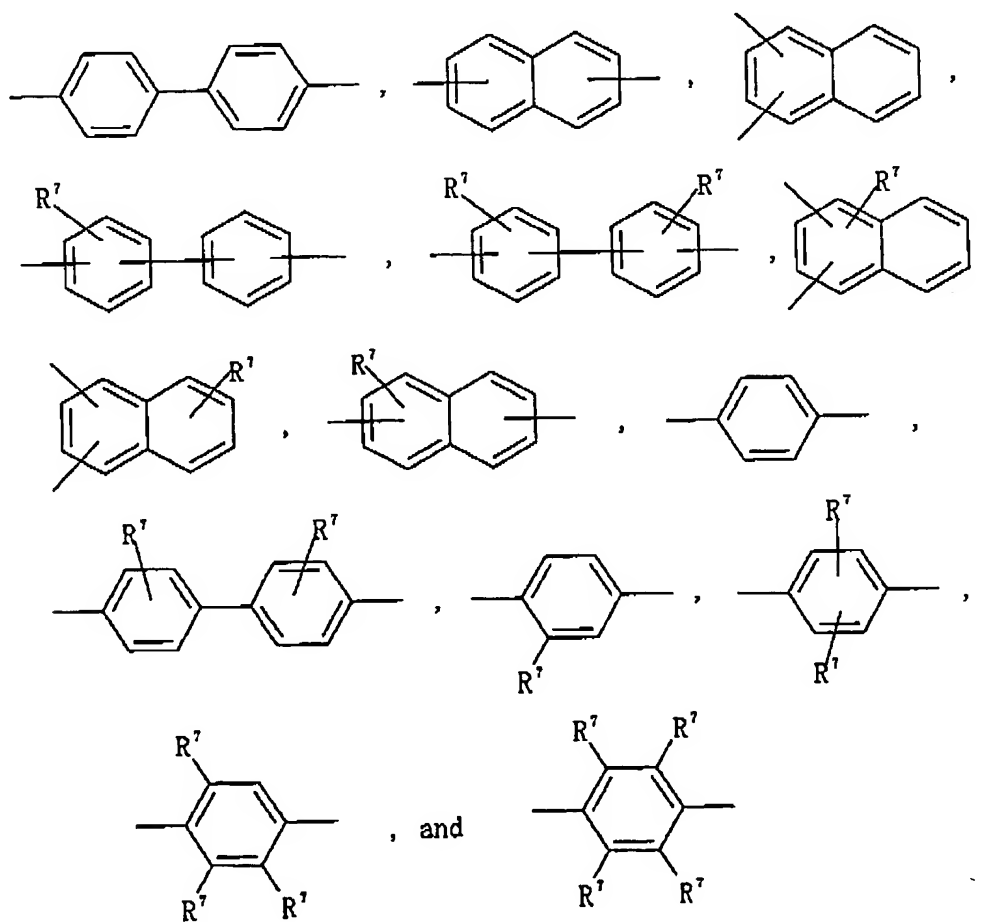
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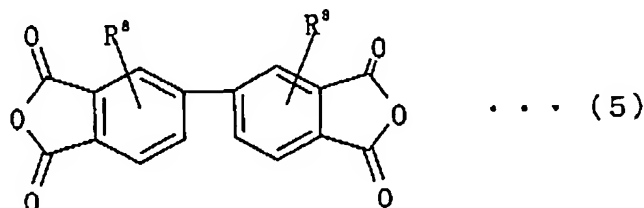
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and each R<sup>7</sup> is independently any one of -H, -CH<sub>3</sub>, -OH, -CF<sub>3</sub>, -SO<sub>4</sub>, -COOH, and -CO-NH<sub>2</sub>, and  
a biphenyl tetracarboxylic dianhydride represented by General Formula (5),



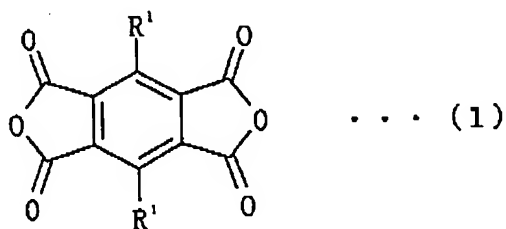


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where  $R^8$  is a residue selected from a group consisting of H-,  $CH_3$ -, Cl-, Br-, F- and  $CH_3O$ -, and  $R^8$  may be the same residues or the different residues,

the polyimide film having a dynamic viscoelasticity whose  $\tan \delta$  peak is located in a range of not less than  $310^\circ\text{C}$  but not more than  $410^\circ\text{C}$ , and whose  $\tan \delta$  value at  $300^\circ\text{C}$  is not more than 0.05.

29. (New) A method in which a pyromellitic dianhydride represented by General Formula (1),



where  $R^1$  is a residue selected from a group consisting of H-,  $CH_3$ -,  $CF_3$ -, Cl-, Br-, F-, and  $CH_3O$ -, and  $R^1$  may be the same residues or different residues,

a paraphenylene diamine represented by General Formula (2)



where  $R^2$  is a bivalent aromatic group selected from a group consisting of:

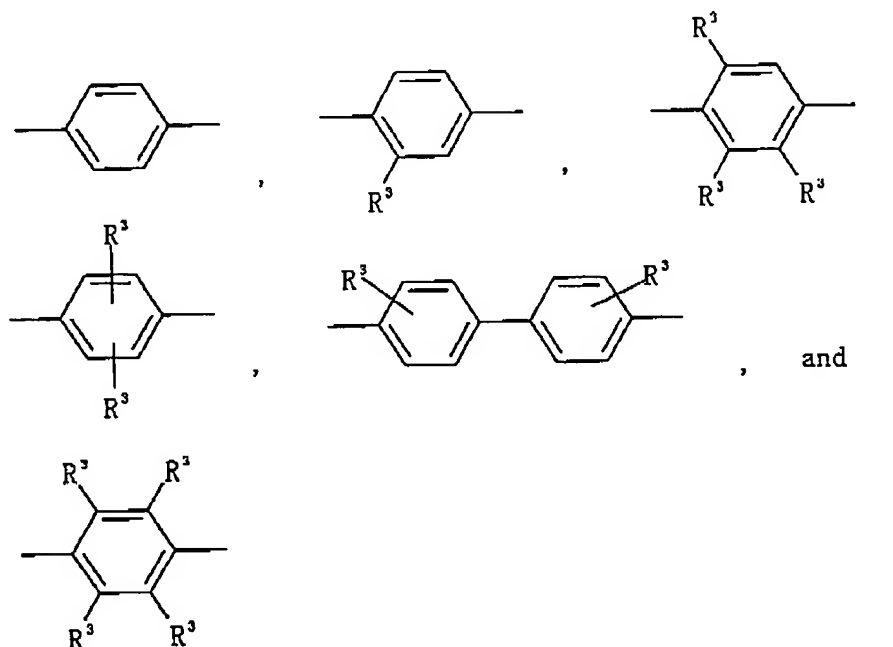
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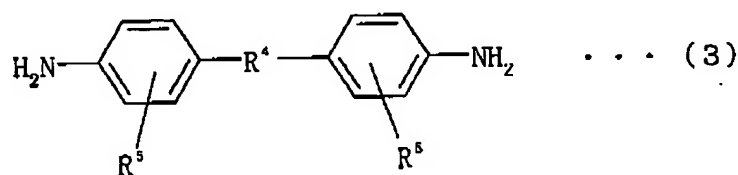
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and each R<sup>3</sup> in the group is independently any one of -H, -CH<sub>3</sub>, -OH, -CF<sub>3</sub>, -SO<sub>4</sub>, -COOH, -CO-NH<sub>2</sub>, -Cl, -Br, -F, and -OCH<sub>3</sub>, and a diaminodiphenyl ether represented by General Formula (3),



where R<sup>4</sup> is a bivalent organic group selected from a group consisting of:

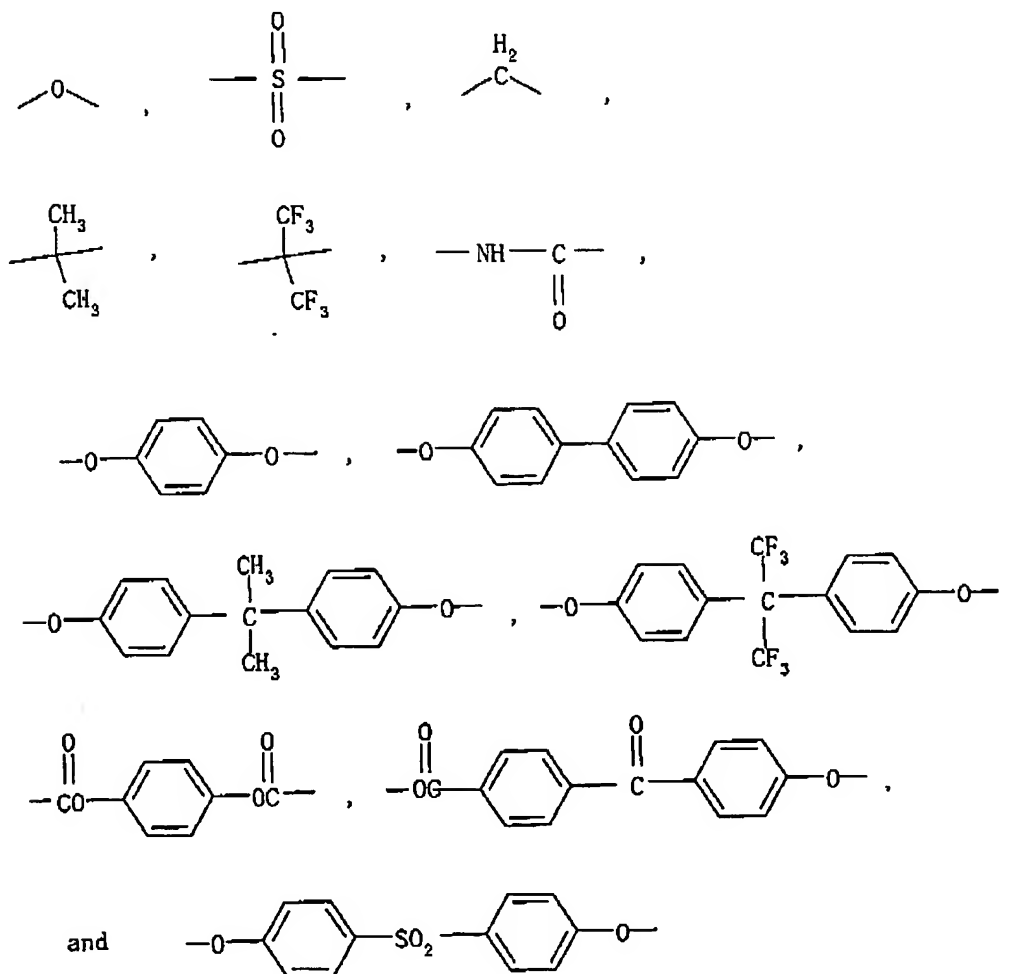
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and each  $R^5$  in the group is independently any one of  $-H$ ,  $-CH_3$ ,  $-OH$ ,  $-CF_3$ ,  $-SO_4$ ,  $-COOH$ ,  $-CO-NH_2$ ,  $-Cl$ ,  $-Br$ ,  $-F$ , and  $-OCH_3$ ,

are co-polymerized as necessary components to manufacture a polyimide film, wherein 5 mole% to 50 mole% of p-phenylene bis(trimellitic monoester anhydride) is used as an acid dianhydride component, so that a peak of  $\tan \delta$  in measuring dynamic viscoelasticity of the polyimide film is controlled in a range of  $310^\circ\text{C}$  to  $410^\circ\text{C}$ .

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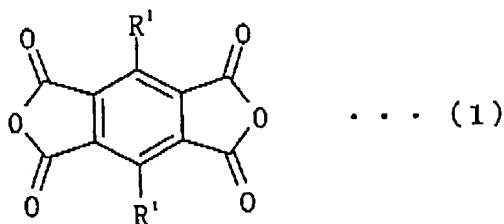
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30. (New) A method for manufacturing a polyimide film, in which a pyromellitic dianhydride represented by General Formula (1),



where R¹ is a residue selected from a group consisting of H-, CH₃-, CF₃, Cl-, Br-, F-, and CH₃O-, and R¹ may be the same residues or different residues,

a paraphenylene diamine represented by General Formula (2)



where R² is a bivalent aromatic group selected from a group consisting of:

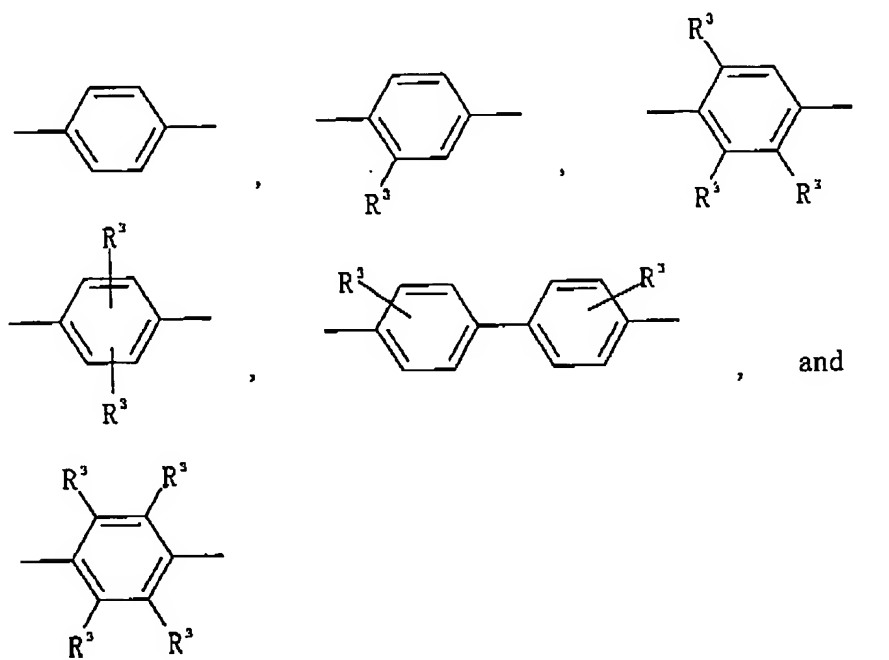
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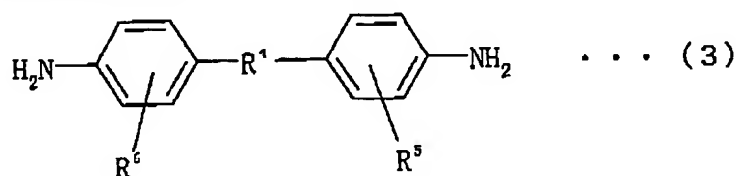
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and each  $R^3$  in the group is independently any one of  $-H$ ,  $-CH_3$ ,  $-OH$ ,  $-CF_3$ ,  $-SO_4$ ,  $-COOH$ ,  $-CO-NH_2$ ,  $-Cl$ ,  $-Br$ ,  $-F$ , and  $-OCH_3$ , and

a diaminodiphenyl ether represented by General Formula (3),



where  $R^4$  is a bivalent organic group selected from a group consisting of:

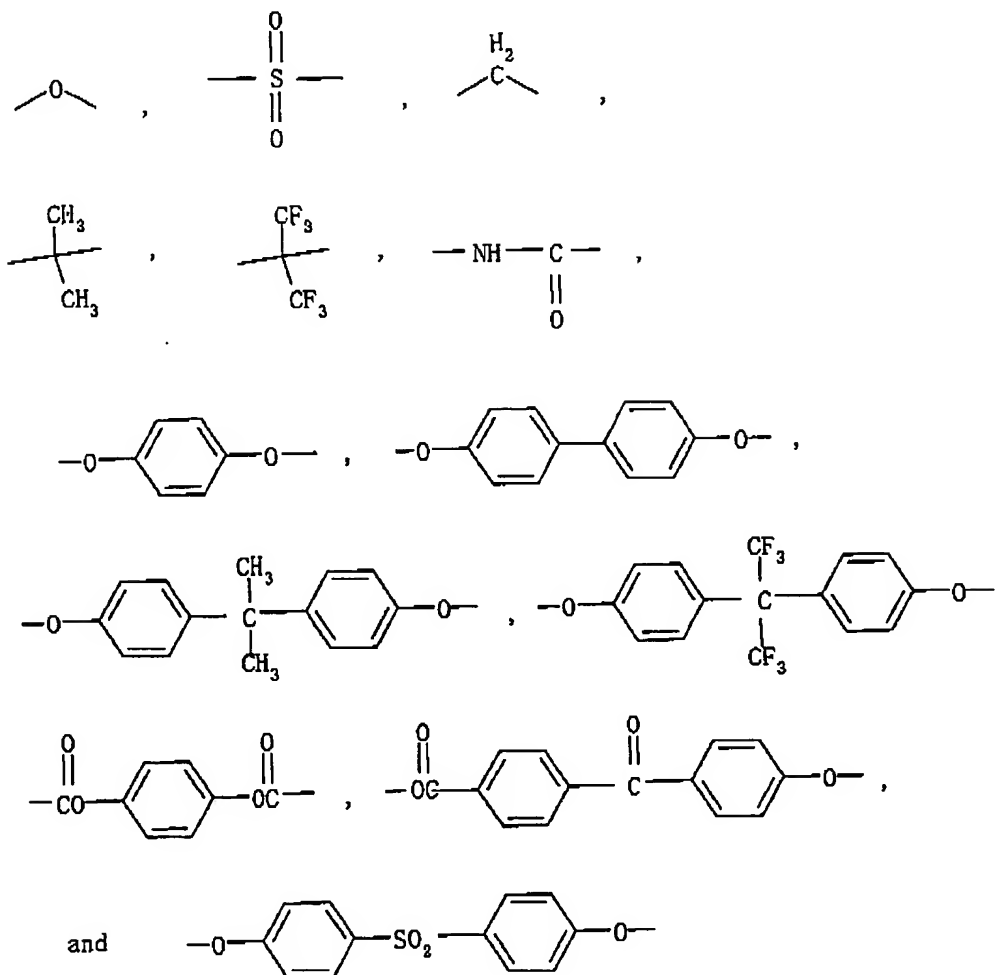
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and each  $R^5$  in the group is independently any one of  $-H$ ,  $-CH_3$ ,  $-OH$ ,  $-CF_3$ ,  $-SO_4$ ,  $-COOH$ ,  $-CO-NH_2$ ,  $-Cl$ ,  $-Br$ ,  $-F$ , and  $-OCH_3$ ,

are co-polymerized as necessary components to manufacture a polyimide film, wherein 5 mole% to 50 mole% of p-phenylene bis(trimellitic monoester anhydride) is used as an acid dianhydride component, and

wherein a peak of  $\tan \delta$  in measuring dynamic viscoelasticity of the polyimide film is controlled in a range of  $310^\circ\text{C}$  to  $410^\circ\text{C}$ .

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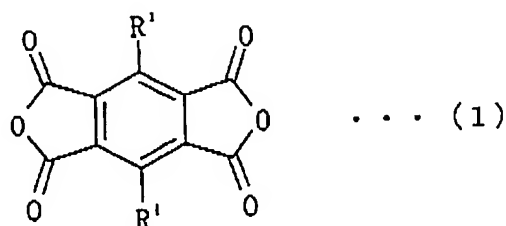
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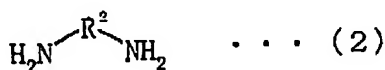
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31. (New) A polyimide film in which a pyromellitic dianhydride represented by General Formula (1),



where R¹ is a residue selected from a group consisting of H-, CH₃-, CF₃-, Cl-, Br-, F-, and CH₃O-, and R¹ may be the same residues or different residues,  
a paraphenylene diamine represented by General Formula (2)



where R² is a bivalent aromatic group selected from a group consisting of:

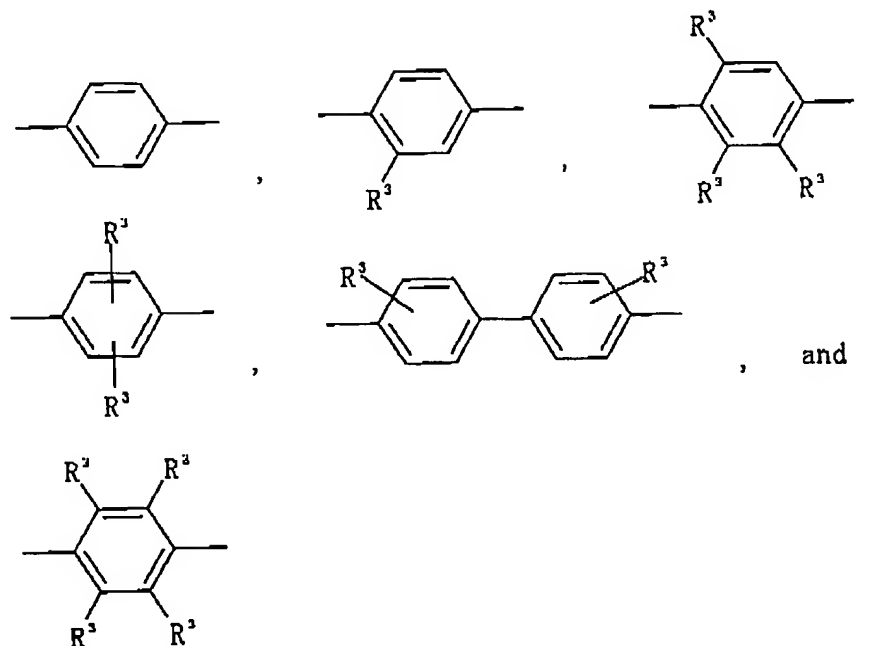
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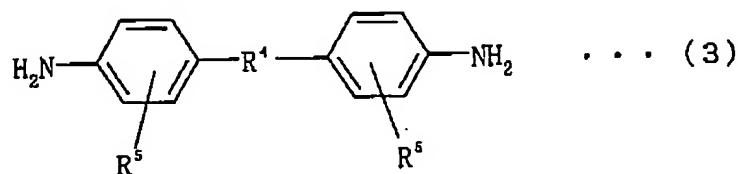
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and each R<sup>3</sup> in the group is independently any one of -H, -CH<sub>3</sub>, -OH, -CF<sub>3</sub>, -SO<sub>4</sub>, -COOH, -CO-NH<sub>2</sub>, -Cl, -Br, -F, and -OCH<sub>3</sub>,  
and a diaminodiphenyl ether represented by General Formula (3),



where R<sup>4</sup> is a bivalent organic group selected from a group consisting of:



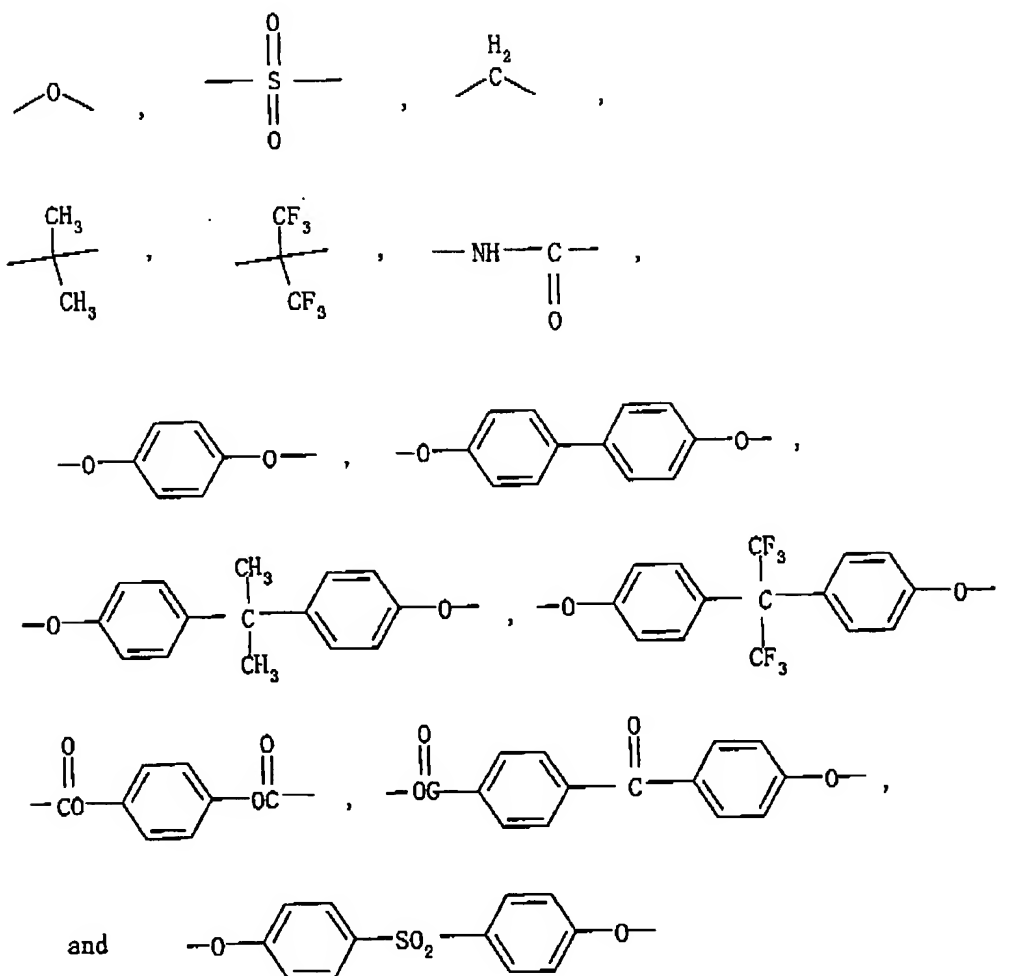
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and each  $R^5$  in the group is independently any one of -H, -CH<sub>3</sub>, -OH, -CF<sub>3</sub>, -SO<sub>4</sub>, -COOH, -CO-NH<sub>2</sub>, -Cl, -Br, -F, and -OCH<sub>3</sub>,

are co-polymerized as necessary components,

the polyimide film being manufactured by a method in which 5 mole% to 50 mole% of p-phenylene bis(trimellitic monoester anhydride) is used as an acid dianhydride component, and in which a peak of tan  $\delta$  in measuring dynamic viscoelasticity of the polyimide film is controlled in a range of 310°C to 410°C.